

# Kondo excitons in self-assembled quantum dots

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## Abstract

We describe novel excitons in quantum dots by allowing for an interaction with a Fermi sea of electrons. We argue that these excitons can be realized very simply with self-assembled quantum dots, using the wetting layer as host for the Fermi sea. We show that a tunnel hybridization of a charged exciton with the Fermi sea leads to two striking effects in the optical spectra. First, the photoluminescence lines become strongly dependent on the vertical bias. Secondly, if the exciton spin is nonzero, the Kondo effect leads to peculiar photoluminescence line shapes with a line width determined by the Kondo temperature.

The Kondo effect concerns the interaction of a localized spin with a Fermi sea of electrons [1]. It is a crucial effect in various areas of nanophysics where often a nano-sized object with spin is in close proximity to a metal [2]. Semiconductor nanostructures are ideal for investigations of Kondo physics because, in contrast to metals, their properties are voltage-tunable [3]. Characteristic to the Kondo effect are a screening of the localized spin and a resonance in the quasi-particle density of states (DOS) at the Fermi energy. This leads to a maximum in conductance at very low temperatures in the transport through a spin- $\frac{1}{2}$  quantum dot [4]. So far, the Kondo effect in nanostructures has been studied almost exclusively in relation to transport properties. In optics, Kondo effects have only been discussed theoretically with respect to nonlinear and shake-up processes in a quantum dot [5, 6].

We present here the theory of novel effects which arise from a combination of the Kondo effect and the optics of nanostructures. In our model, a charged exciton in a quantum dot (QD) interacts with a Fermi sea of free electrons. We call the resulting quasi-particles Kondo-excitons. We suggest that such novel excitons can exist in self-assembled quantum dots where charged excitons can be prepared in voltage-tunable structures [7]. Furthermore, the wetting layer can be filled with electrons and used as a two-dimensional (2D) electron gas (fig. 1a), offering a simple means of generating a Fermi sea in close proximity to a quantum dot. A significant point is that self-assembled dots are very small, only a few nanometers in diameter, allowing the Kondo temperature  $T_K$  to be as high as  $\sim 10$  K. We find that the photoluminescence (PL) from Kondo excitons is determined by the Kondo DOS at the Fermi level such that the PL line width is equal to  $k_B T_K$  ( $k_B$  is the Boltzmann constant). Furthermore, the PL energy depends strongly on the Fermi energy, and therefore also on a bias voltage. This behavior contrasts with conventional single dot PL where the lines are very sharp depending only weakly through the Stark effect on the bias [7].

In voltage-tunable structures, self-assembled QDs are embedded between two contacts [7, 8]. This makes it possible to control the number of electrons in a QD by application of a voltage  $U_g$  (fig. 1a) [8]. By generating a hole with optical excitation, it has been demonstrated that there are regions of gate voltage with constant excitonic charge, with the excitonic charge changing abruptly at particular values of  $U_g$  [7]. The charged excitons, labelled  $X^{n-}$ , contain  $n + 1$  electrons and one hole. Excitons with  $n$  up to 3 have been observed in InAs/GaAs quantum dots [7, 9]. At higher voltages, capacitance-voltage spectroscopy shows that electrons fill the 2D wetting layer [8]. In this case, the Fermi energy ( $E_F$ ) in the wetting layer depends linearly on  $U_g$ : we find that  $E_F = (a_o^*/4d)(U_g - U_g^o)$ , where  $a_o^*$  is the effective Bohr radius,  $d$  is the distance between the back gate and wetting layer,  $U_g^o$  is the threshold gate voltage at which electrons start to fill the wetting layer, and  $d \gg a_o^*$  [8]. Existing PL results in the regime of wetting layer filling show broadenings and shifts of the PL lines [7], which we believe is some evidence of an interaction with the Fermi sea, although this has not been analyzed in detail.

Typically, the QD  $X^0$  and  $X^{1-}$  excitons are strongly bound and exist only at gate voltages

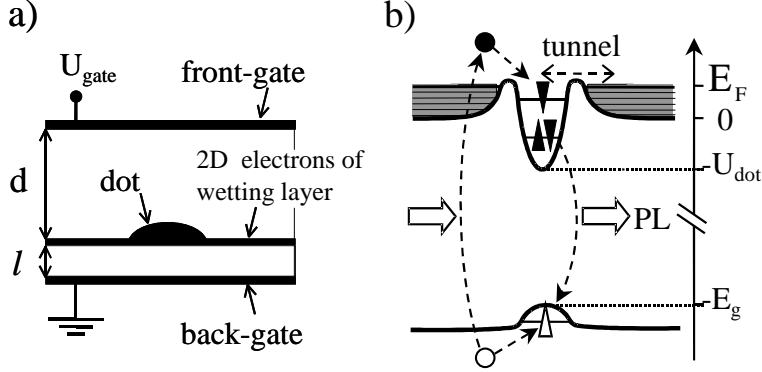


FIG. 1: (a) Schematic of the heterostructure with a quantum dot embedded between front and back gates;  $d \ll l$ . (b) The band diagram of a quantum dot and associated wetting layer showing also the energies typical to self-assembled quantum dots.

where the wetting layer is empty. It is therefore unlikely that  $X^0$  and  $X^{1-}$  can couple with the Fermi sea. Instead, we focus on the excitons  $X^{2-}$  and  $X^{3-}$  for which the electrons on the upper state may couple with extended states of the Fermi sea as we illustrate in fig. 1b. Since self-assembled QDs are usually anisotropic, we model a QD with two bound non-degenerate orbital states, having indexes  $s$  and  $p$ . For a  $p$  state electron, the electrostatic potential consists of the QD short-range confinement and the long-range Coulomb repulsion from the charges in the lower  $s$  state. This potential will therefore have a barrier at the edge of the QD (fig. 1a), allowing the  $p$  electron(s) to tunnel in and out of the QD.

To investigate the interaction between a quantum dot exciton and a Fermi sea of electrons, we employ the Anderson Hamiltonian which includes the single-particle energy  $\hat{H}_{sp}$ , the intra-dot Coulomb interaction, and a hybridization term  $\hat{H}_{tun}$ :

$$\hat{H} = \hat{H}_{sp} + \frac{1}{2} \sum_{\alpha_1, \alpha_2, \alpha_3, \alpha_4} U_{\alpha_1, \alpha_2, \alpha_3, \alpha_4}^{ee} a_{\alpha_1}^+ a_{\alpha_2}^+ a_{\alpha_3} a_{\alpha_4} - \sum_{\alpha_1, \alpha_2, \alpha_3, \alpha_4} U_{\alpha_1, \alpha_2, \alpha_3, \alpha_4}^{eh} a_{\alpha_1}^+ b_{\alpha_2}^+ b_{\alpha_3} a_{\alpha_4} + \hat{H}_{tun} \quad (1)$$

where  $a_{\alpha}^+$  ( $b_{\alpha}^+$ ) is the intra-dot creation operator of electrons (holes);  $U^{ee}$  and  $U^{eh}$  are electron-electron and electron-hole Coulomb potential matrix elements, respectively. The index  $\alpha$  stands for  $(\beta, \sigma)$ , where  $\beta$  is the orbital index and  $\sigma$  the spin index;  $\beta$  can be  $s$  or  $p$ , and  $\sigma = \pm \frac{1}{2}$  for electrons and  $\pm \frac{3}{2}$  for heavy holes.  $\hat{H}_{tun}$  is given by:  $\hat{H}_{tun} = \sum_{\sigma} V_k [c_{k, \sigma}^+ a_{p, \sigma} + a_{p, \sigma}^+ c_{k, \sigma}]$ , where the operators  $c_{k, \sigma}$  describe the delocalized electrons in the Fermi sea;  $k$  and  $E_k$  are the 2D momentum and kinetic energy, respectively. For the tunnel matrix element  $V_k$ , we assume  $V_k = V$  in the interval  $0 < E_k < D$  and  $V_k = 0$  elsewhere [10]. In the operator  $\hat{H}_{tun}$ , we include only coupling between the  $p$  state and the Fermi sea. In our approach,

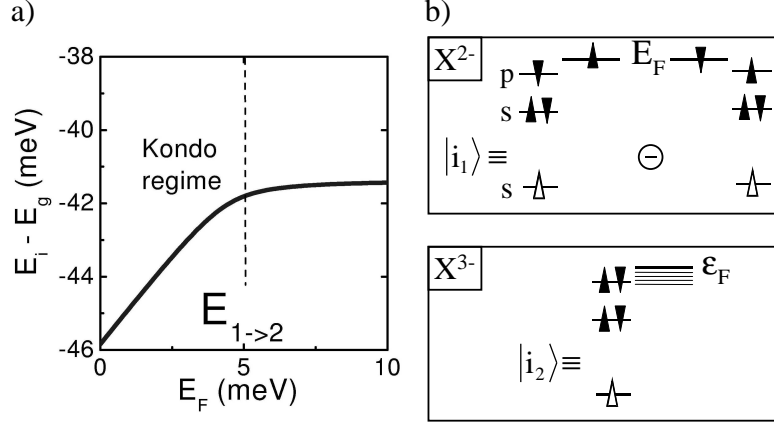


FIG. 2: (a) Calculated energy of the initial state as a function of the delocalized state energy. The energy  $E_F$  plays the role of the Fermi level. Here,  $U_{dot} = 64$  meV and  $E_g$  is the band gap of the QD. (b) Electron configurations contributing to the initial ground state. Electron (hole) spins are represented with solid (open) triangles.

the quantization in a QD is assumed to be strong so that the Coulomb interactions can be included with perturbation theory [11, 12]. The tunnel broadening is the smallest energy in the problem. Our approach is to solve eq. 1 for the initial and final states, and calculate the optical emission spectrum at zero temperature:

$$I(\omega) = Re \int_0^\infty dt e^{-i\omega t} \langle i | \hat{V}_o^+(t) \hat{V}_o(0) | i \rangle,$$

where  $|i\rangle$  is the initial state and the operator  $\hat{V}_o = V_{opt}(b_{s,-\frac{3}{2}}a_{s,\uparrow} + b_{s,\frac{3}{2}}a_{s,\downarrow} + c.c.)$  describes the strong transitions between the hole and electron  $s$  states.

We start with a zero bandwidth model, in which the Fermi sea is replaced by a single “delocalized” state of energy  $E_F$  [1]. In this simplified model,  $E_F$  plays the role of the Fermi level allowing us to predict the main features in the evolution of the PL with Fermi energy. We describe the electron wave function with the occupations of the electron states, labelling the delocalized state  $e$ . The aim is explore the hybridization of the  $X^{2-}$  and  $X^{3-}$  excitons, so we consider 4 electrons and one hole. We take the hole angular momentum as  $+\frac{3}{2}$  (denoted  $s_{+\frac{3}{2}}$ ); equivalent results are obtained also with  $-\frac{3}{2}$ . The electron wave function of the hybridized initial state before photon emission will have total electron spin  $S_e = 0$ . We therefore express the ground state as  $|i\rangle = A_1|i_1\rangle + A_2|i_2\rangle$ , where:

$$\begin{aligned} |i_1\rangle &= \frac{1}{\sqrt{2}}(|s_\uparrow, s_\downarrow, p_\uparrow, e_\downarrow\rangle - |s_\uparrow, s_\downarrow, p_\downarrow, e_\uparrow\rangle)|s_{+\frac{3}{2}}\rangle, \\ |i_2\rangle &= |s_\uparrow, s_\downarrow, p_\uparrow, p_\downarrow\rangle |s_{+\frac{3}{2}}\rangle. \end{aligned}$$

The states  $|i_1\rangle$  and  $|i_2\rangle$  correspond to the excitons  $X^{2-}$  and  $X^{3-}$ , respectively (fig. 2), and have energies  $E_{i_1}$  and  $E_{i_2}$ . In the Kondo function  $|i_1\rangle$ , the delocalized electron “screens” the net spin in the QD. By diagonalising the Hamiltonian, we find that the initial ground state has energy  $E_i = \frac{1}{2}(E_{i_1} + E_{i_2} - \sqrt{(E_{i_1} - E_{i_2})^2 + 8V^2})$  and that  $A_1 = -a/(1 + a^2)^{1/2}$ ,  $A_2 = -A_1/a$ , where  $a = (E_{i_2} - E_i)/\sqrt{2}V$ .

As the energy  $E_F$  increases, the ground state evolves from the  $X^{2-}$  to the  $X^{3-}$  exciton. The transition occurs when  $E_{i_1} \approx E_{i_2}$  where  $E_F \approx E_{1\rightarrow 2} = E_2^{\text{intra}} - E_1^{\text{intra}}$  where  $E_1^{\text{intra}}$  and  $E_2^{\text{intra}}$  are the intra-dot energies. The hybridization between  $|i_1\rangle$  and  $|i_2\rangle$  corresponds to the so-called mixed-valence regime [1]. To calculate  $E_i$  numerically, we represent a QD as an anisotropic harmonic oscillator taking the electron (hole) oscillator frequencies as 25 and 20 (12.5 and 10) meV. Fig. 2a shows  $E_i$  as a function of  $E_F$ .

After photon emission from state  $|i\rangle$ , the final state  $|f\rangle$  has  $S_e = \frac{1}{2}$ . The intra-dot configuration in the  $X^{2-}$  final state is either a *singlet* ( $S_{\text{dot}} = 0$ ), with configuration  $\frac{1}{\sqrt{2}}(|s_\uparrow, p_\downarrow\rangle - |s_\downarrow, p_\uparrow\rangle)$ , or a *triplet* ( $S_{\text{dot}} = 1$ ), with configurations  $|s_\uparrow, p_\uparrow\rangle$  and  $\frac{1}{\sqrt{2}}(|s_\uparrow, p_\downarrow\rangle + |s_\downarrow, p_\uparrow\rangle)$  [7] (Fig. 3a). The PL related to these configurations are well separated, typically by  $\sim 5$  meV [7, 9], due to the exchange interaction between the  $s$  and  $p$  states. Non-zero optical matrix elements exist for the three final states:

$$\begin{aligned} |f_1\rangle &= \frac{1}{\sqrt{6}}(|s_\uparrow, p_\downarrow, e_\uparrow\rangle + |s_\downarrow, p_\uparrow, e_\uparrow\rangle - 2|s_\uparrow, p_\uparrow, e_\downarrow\rangle), \\ |f_2\rangle &= \frac{1}{\sqrt{2}}(|s_\uparrow, p_\downarrow, e_\uparrow\rangle - |s_\downarrow, p_\uparrow, e_\uparrow\rangle), \\ |f_3\rangle &= |s_\uparrow, p_\uparrow, p_\downarrow\rangle. \end{aligned}$$

In the state  $|f_1\rangle$ , the intra-dot triplet state with  $S_{\text{dot}} = 1$  is “screened” by the delocalized electron.  $|f_2\rangle$  has the singlet intra-dot state, and the function  $|f_3\rangle$  plays the main role in the emission of the  $X^{3-}$  exciton.

The calculated PL spectrum contains three lines (Fig. 3), labelled as  $X_t^{2-}$ ,  $X_s^{2-}$  and  $X^{3-}$ , with relative intensities  $\frac{3}{4}A_1^2 : \frac{1}{4}A_1^2 : A_2^2$ . Without the tunnel interaction, there is an abrupt jump in the PL from  $X^{2-}$  to  $X^{3-}$ , but with the tunnel interaction, the PL shows very clearly hybridization effects. In particular, in the mixed-valence regime  $E_F \approx E_{1\rightarrow 2}$ , the intensity of the  $X^{2-}$  lines rapidly decrease and the  $X^{3-}$  line appears but the energies of all three lines depend on the delocalized state energy  $E_F$ . Hence, a clear prediction is that in the hybridization regime, the PL depends on the Fermi energy and therefore also on the gate voltage.

We focus now on the PL line shapes in the Kondo regime  $E_F < E_{1\rightarrow 2}$ . While the zero bandwidth model is adequate for the general picture of the PL, we have to employ the *finite* bandwidth model in order to calculate the PL line shapes. The initial Kondo state is the exciton  $X^{2-}$  coupled with the Fermi sea. A trial function for the  $S_e = 0$  ground state is [1, 13]:

$$|i\rangle = [A_0|\phi_0\rangle + \sum_{k>k_F} A_k|\phi_k\rangle] * |s_{+\frac{3}{2}}\rangle; \quad (2)$$

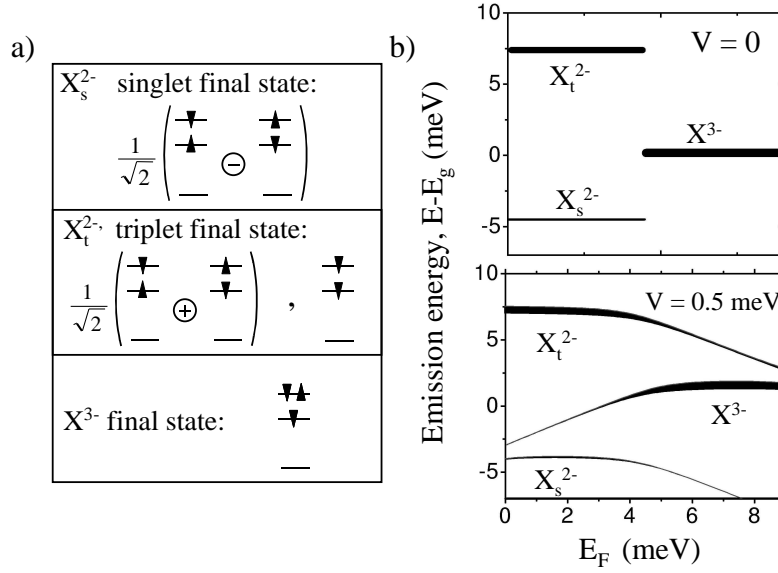


FIG. 3: (a) Final intra-dot states related to the singlet and triplet states of the  $X^{2-}$  exciton, and to the  $X^{3-}$  exciton. (b) Calculated energies of the optical transitions as a function of the delocalized state energy without ( $V = 0$ ) and with ( $V = 0.5$  meV) the hybridization. The thickness of the line represents the intensity of the emission.

$$|\phi_0\rangle = |s_\uparrow, s_\downarrow, p_\uparrow, p_\downarrow; \Omega\rangle,$$

$$|\phi_k\rangle = \frac{1}{\sqrt{2}} \left( \hat{c}_{k,\downarrow}^\dagger |s_\uparrow, s_\downarrow, p_\uparrow; \Omega\rangle - \hat{c}_{k,\uparrow}^\dagger |s_\uparrow, s_\downarrow, p_\downarrow; \Omega\rangle \right),$$

as represented diagrammatically in Fig. 4a. The configurations are described with the localized states and with the symbol  $\Omega$  which denotes all states of the Fermi sea with  $k < k_F$ , where  $k_F$  is the Fermi momentum. From the Anderson Hamiltonian, we find that [1]:

$$A_k = -\frac{\sqrt{2}VA_0}{E_1^{\text{intra}} + E_k - E_i}, \quad A_0 = \frac{1}{(1 + 2\Delta/\pi\delta)^{1/2}},$$

where  $\Delta = \pi V^2 \rho$  ( $\rho$  is the 2D DOS). We write the ground state energy as  $E_i = E_1^{\text{intra}} + E_F - \delta$ , where  $\delta$  is the lowering of energy due to the Kondo effect. If  $\delta < E_{1-2} - E_F$ , we obtain

$$\delta = (D - E_F) \exp \left( -\frac{\pi(E_2^{\text{intra}} - E_1^{\text{intra}} - E_F)}{2\Delta} \right) = k_B T_K. \quad (3)$$

Here, the energy  $\delta$  plays the role of the Kondo temperature in the initial state,  $T_K$ . The temperature  $T_K$  can be as high as  $\sim 7 - 14$  K for realistic parameters  $E_F = 2 - 3.6$  meV,  $\Delta = 1$  meV, and  $D \sim 30$  meV.

In the regime  $E_F < E_{1-2}$ , the final *intra-dot* configuration is either a singlet or a triplet state (fig. 3a), as in the case of the zero bandwidth calculation. The triplet state couples with the Fermi gas, leading to a state with total electron spin  $S_e = \frac{1}{2}$ . However, the Kondo

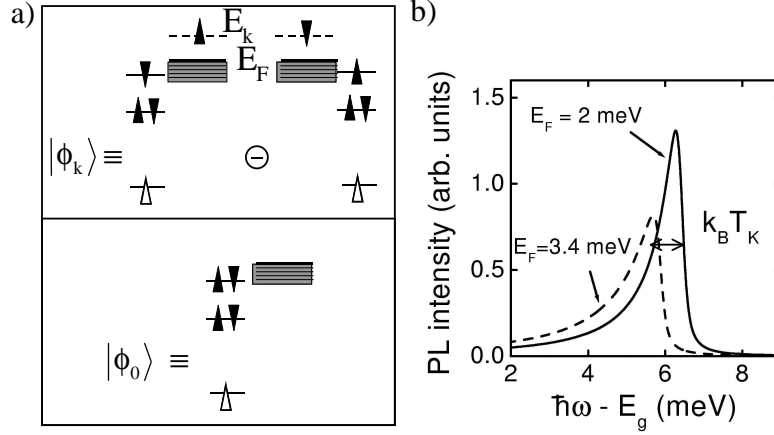


FIG. 4: (a) Contributions to the initial state (2); (b) Calculated emission spectrum of the  $X_t^{2-}$  exciton with the triplet final state configuration;  $\Delta = 1$  meV. The Fermi energies of 2 and 3.6 meV correspond to  $KT_K = 7$  and 14 K, respectively.

temperature for this final state turns out to be much less than  $T_K$  allowing us to use the single-particle final states to calculate the PL spectrum. The singlet final state, and also the  $X_t^{3-}$  final state, have higher energy and are broadened by energy  $\Delta$  through tunnelling into empty delocalized states. From state  $|i\rangle$  in eq. 2, we write the spectral function  $I(\omega)$  in the form:

$$I(\omega) = -Re[i \sum_{\beta, \beta'} A_{\beta} A_{\beta'} F_{\beta, \beta'}],$$

where  $\beta$  can be either 0 or  $k$ ,  $F_{\beta\beta'} = \langle \phi_{\beta} | \hat{V}_o^+ \hat{R} \hat{V}_o | \phi_{\beta'} \rangle$ , and  $\hat{R} = 1/(\hat{H} + \hbar\omega - E_i - i0)$ . We find that the  $X_t^{2-}$  PL line has an asymmetric shape (Fig. 4):

$$X_t^{2-}(\omega) = V_{opt}^2 \frac{3\Delta A_0^2}{2\pi} \int_0^{D-E_F} d\epsilon \frac{1}{(\epsilon + k_B T_K)^2} Re \frac{-i}{\hbar\omega - E'(X_t^{2-}) + \epsilon - i\gamma}, \quad (4)$$

where  $E'(X_t^{2-}) = E(X_t^{2-}) - k_B T_K$  is the renormalized emission energy and  $\gamma$  describes the broadening of the final state. We can expect  $\gamma$  to be small since the relaxation of the final state requires a spin-flip [7], and in this case,  $k_B T_K \gg \gamma_t$ , PL line width is equal to  $KT_K$ . In other words, the PL reflects the spectral DOS near the Fermi level in the initial Kondo state.

The important result is that in the Kondo regime, both the PL peak position and the PL line shape depend on the Fermi energy and, hence, on the gate voltage. Furthermore, as the temperature increases, the peak in the spectral DOS diminishes rapidly [1] and therefore the line width of the Kondo-exciton  $X_t^{2-}$  should also be strongly temperature dependent.

In the general case of arbitrary  $E_F$ , the finite bandwidth model leads to the PL spectrum similar to that in fig. 3b [14]. The  $X_s^{2-}$  and  $X^{3-}$  PL lines are close to Lorentzians, with line widths of  $\Delta$  due to the finite lifetime of the final states. It is important to emphasize that the  $X_t^{2-}$  final state lies at lower energy and does not suffer from tunnel broadening such that the line shape and line width are determined by many-body effects.

In summary, we have described novel excitons which can exist in self-assembled QDs when there is an interaction between a charged exciton localized on the dot and delocalized electrons in the wetting layer. The charged exciton with nonzero spin is surrounded by a “cloud” (of radius  $\propto (k_B T_K)^{-1}$ ) of Fermi electrons. We predict several striking manifestations of these novel states in the emission spectra: at low temperature, the optical lines are strongly dependent on vertical bias and their widths are determined by the Kondo temperature.

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